



Development of ultraviolet laser for disinfection of potable water

G C Bhar* and U Chatterjee

Laser Laboratory, Physics Department, Burdwan University, Burdwan-713 104, West Bengal, India

E-mail : dgp_buphysch@sanchernet.in

Abstract : The ultraviolet (UV) radiation is an effective means against bacteria and protozoan pathogens for disinfection of drinking water killing micro-organisms by causing irreparable damage to their DNA. Some protozoan cysts like giardia require a high dose of UV intensity of $16000 \mu\text{W.s/cm}^2$ for total destruction. Low level of UV radiation as that available from commercial source for insecticides and viricide is incapable of elimination of such resistive cysts. So UV radiation is of demand for this. But except excimer lasers that are of limited life and hard to operate, there is practically no UV laser as such. The easy way is to generate through nonlinear frequency mixing of laser radiations from Nd : YAG and Dye lasers in a suitable nonlinear crystal. Except borates, few nonlinear crystals can be used in the UV region down to 200 nm or below. The borate group includes BBO, LBO, CLBO, LB_4 etc. while the most recent addition is KABO. Although the generation of UV radiation of 240–280 nm that is important for DNA absorption is of no problem, but tuning below 200 nm is not quite easy. The shortest wavelength we have generated is 187.9 nm in BBO through sum frequency mixing. We have shown that it can be tuned to still lower wavelength if impurity-free BBO is used and the crystal be cooled. Crystal LB_4 is better in this respect as it transmits down to 170 nm. But since nonlinearity of LB_4 is very low, available intensity of UV radiation may not be sufficient for such disinfecting applications. So some ingenious technique of multipass has been proposed and demonstrated to realize high conversion in such crystals.

Keywords : UV radiation, nonlinear optical devices, frequency mixing, multipass configuration.

PACS Nos. : 42.65.Ky, 42.72.Bj

1. Introduction

The ultraviolet (UV) radiation is an effective means against bacteria and protozoan pathogen for disinfection of drinking water. And in fact it is better than other disinfectants like ozone, chlorine, chlorine dioxide and chloramine as it does not leave harmful residue. The UV radiation kills microorganism by causing irreparable damage to their DNA.

There are four principal factors that need discussion determining effectiveness of UV light inactivation of microorganism :

- (i) Wavelength of UV radiation,
 - (ii) Intensity of UV radiation,
 - (iii) Exposure time of UV radiation,
 - (iv) Resistance of target microorganism.
- The wavelength of interest for disinfection is 250–270 nm. The commercial sources include some incandescent

light bulbs, halogen bulbs, high efficiency light bulbs and also computer screen which also emit UV radiation that are too weak for this purpose. However, special UV emitting lamps are used to disinfect water. Commercially available low-pressure mercury vapour lamps emit principal UV wavelength at 254 nm. Maximum dose of many commercially available UV units is $25\text{--}35 \text{ mW/cm}^2$. Typical UV intensity requirements for disinfection studied in advanced countries [1] are the following :

- (a) Bacteria : E coli – 3 mJ/cm^2 ,
Vibro Cholerae – 3.4 mJ/cm^2 .
- (b) Viruses : Hepatitis A virus – 20 mJ/cm^2 .
- (c) Protozoan cysts : Giardia muris – 82 mJ/cm^2 .

As a result, one of the world's largest potable disinfection plants in London uses UV as the primary disinfectant and adds free chlorine for the residual infectants prior to storage. Since using commercial

*Corresponding Author

nonlaser UV source, famous M/S WaterHealth International of USA clarifies that "it does not treat parasites or microorganism cysts with protective coverings such as *Cryptosporidium* and *Giardia*."

The UV radiation after absorption alters nitrogenous heterocyclic components within DNA and RNA and causes molecules to form new bonds. This effect renders microorganism unable to replicate. The UV dosage measured in mW.sec per sq. centimeter must either eliminate DNA replication or partially damage the genetic material [2]. Under certain conditions, cells damaged by UV may repair and reactivate themselves through enzyme activity. Photoreactivation *i.e.* the revival of organism in visible light shortly after UV light exposure, may be a function of the light intensity. Permanent inactivation requires damage to nucleic acids for which key parameter is an appropriate UV dose. So high intensity UV light as that generated by laser is the answer to this problem.

But there is practically no UV laser as such, excepting the excimer ones which are of limited life and hard to operate. The easy way is to generate through harmonic generation or sum frequency mixing of laser radiation from commercially available Nd : YAG and Dye lasers in a suitable nonlinear crystal. There are not many such nonlinear crystals transmitting in the UV region of 200 nm or below except borates. These include BBO, LBO, CLBO, LB4 and the most recent addition is KABO. The UV generation in the region 240–280 nm, important for DNA absorption, is of no problem. The generation of fourth harmonic at 266 nm of common Nd : YAG laser line has been realized in CLBO crystal. Tuning below 200 nm is difficult and the shortest wavelength we have generated is 187.9 nm in BBO through sum frequency mixing (SFM). Crystal LB4 is better in this respect but since its nonlinearity is very low, available intensity of

UV radiation may not be sufficient for the application. So some ingenious technique of multipass has been proposed and demonstrated to realize high conversion in such crystals.

2. UV generation through frequency-mixing in Nonlinear Crystals

We have been engaged in characterization of various UV and IR crystals meant for different applications. The UV radiations are generated through third harmonic generation (THG) of Nd : YAG laser and Dye laser and also through sum frequency mixing of these two input laser radiations. The techniques are described in crystals of BBO, LBO, CLBO and LB4 and means are demonstrated for efficient conversion into UV since high UV intensity is necessary for removal of resistant microorganism. Out of these, the crystal of most interest is BBO having highest nonlinearity of the lot and its UV transmission is as low as 187 nm. It can still be shifted to shorter wavelength if the impurity content causing onset of band-edge-absorption can be reduced by after-growth quality improvement technique. With the availability of various samples of crystals from BARC, it had been possible to verify the concept [3,4]. Linear and nonlinear characterizations of UV transmitting crystals are illustrated in Table 1.

3. Experiments and results

Our basic pump laser is Spectra Physics model DCR-11 which is a Q-switched Nd : YAG laser having pulse repetition rate of 10 pps while the Dye laser is of the same make (model PDL-2) pumped by the second harmonic of the Nd : YAG laser. The dye laser is capable of operating through the entire visible and very near infrared. The following two types of nonlinear

Table 1. Linear and nonlinear optical parameters of some borate group crystals.

Crystal	BBO	LBO	CBO	CLBO	LB ₄
Symmetry	3 c	3 m	222	42 m	4 mm
Transparency (μm)	0.19–3.4	0.16–2.6	0.17–3.0	0.18–2.75	0.16–3.5
Absorption coefficient (cm^{-1}) at 200 nm	0.96	1.10	0.50	0.0042	0.004
Birefringence	0.11	0.039	0.057	0.0496	0.0546
Effective nonlinear coefficient d_{eff} (pm/V)	2.20	1.20	3.67	0.47	0.16
Damage threshold (1 ns, 10 pps) (GW/cm^2)	13	27	20–26	26	40
Walk-off (degree)	4	0.37	1.76	1.78	2.1
SHG cut-off (nm)	411	554	480	470	487
Chemical property	Hygroscopic	Non-hygroscopic	Hygroscopic	Hygroscopic	Non-hygroscopic

interactions have been studied for generation of UV radiation :

- (i) Harmonic generation of 2nd and 3rd of Nd : YAG laser and Dye laser in CLBO and LB4 crystals.
- (ii) Sum frequency mixing in BBO, CLBO and LBO crystals.

The experimental arrangements along with tuning range for second harmonic generation (SHG) and third harmonic generation (THG) of Dye laser radiations in CLBO are illustrated in Figures 1 and 2 respectively. In the SHG

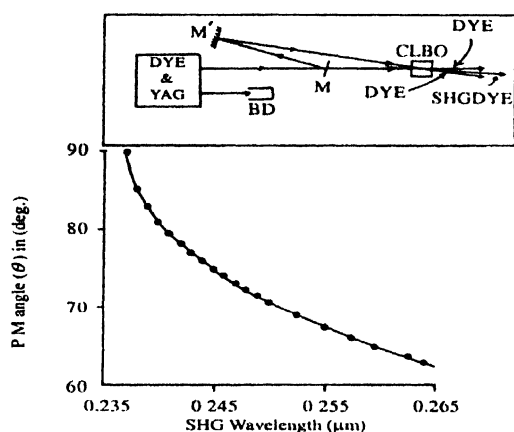


Figure 1. Phase-matching characteristic for collinear SHG (236.8–265 nm) in a CLBO crystal. The experimental points (•) are shown over the theoretical curve (solid line). The inset shows the experimental setup for noncollinear configuration. BD is beam dumper for Nd : YAG laser radiation. M and M' are dichoric mirrors, M being a partially transmitting dichoric mirror.

experiment, frequency doubling of dye laser (74ATFMC) the generated harmonic is 236.8–265 nm. A long focal length (110 cm) lens is used to loosely focus the fundamental dye laser radiation in the CLBO crystal. A dichoric filter is used to separate the residual fundamental beam from the generated UV. The crystal being non-centrosymmetric, the three wave mixing for THG requires a second stage where the residual fundamental beam after SHG is sum-frequency mixed with the SHG in a second nonlinear crystal. Thus for the THG experiment in CLBO, dye (LDS-698) laser radiation producing fundamental 678.9–713 nm beam is mixed with its second harmonic radiation 339.5–356.6 nm made in a Type-I, $\theta = 30^\circ$ cut, 7.3 mm long BBO crystal [5]. Figure 2 shows the phase-matched tuning range 226.3–237.5 nm for different noncollinear angle between the fundamental and harmonic beam. The short wavelength cut-off arises due to noncritical phase-matching while in

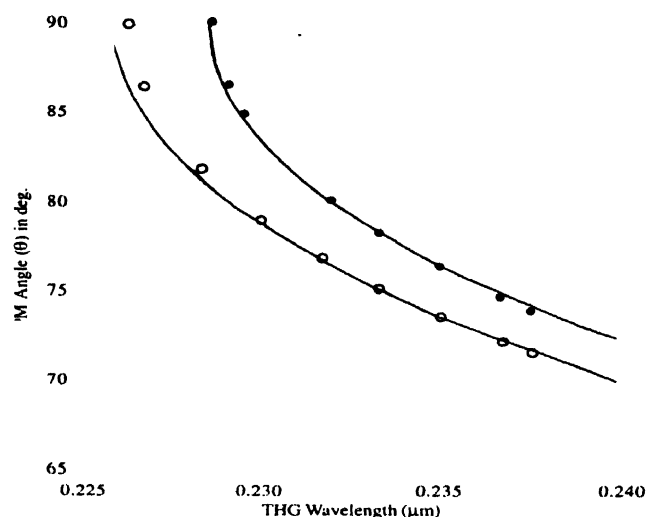


Figure 2. Phase-matching characteristics of CLBO crystal for THG of LDS-698 nm dye laser radiation (678.9–713 nm) by noncollinear configuration for internal noncollinear angle $\alpha = 0.5^\circ$ (lower curve) and 5.7° (upper curve).

our experiment, the long wavelength limit is set by the crystal cut. Hence, the latter can be extended further using another suitably cut CLBO crystal. The region below 230 nm and down to 200 nm is covered by THG in a Type-I, $\theta = 60^\circ$ cut BBO crystal using dye laser radiations, the dyes used being LDS-698 and DCM.

Shorter wavelength region is accessed through sum-frequency mixing in crystals having deep UV transmission. The region below 200 nm has been covered in BBO and CLBO crystals. The experimental arrangements are illustrated in Figure 3. First the second harmonic of a dye (Coumarine) laser covering a tuning range of 456–530 nm is generated in a Type-I, $\theta = 60^\circ$ cut BBO crystal. This is then sum-frequency mixed in a CLBO crystal with fundamental Nd : YAG laser radiation (1064 nm). The interaction is made noncollinear to avoid use of filter for separation of the generated 195.3–210 nm from the input parent beams. The limiting generated wavelength is 195.3 nm employing a noncollinear angle of 8° . In BBO crystal, still shorter wavelength can be accessed by sum-frequency mixing due to its available UV transmission and phase-matchability thereof. To cover the longer wavelength UV generation of 240–280 nm, we have used LBO crystal and noncollinear sum-frequency mixing of Nd : YAG and dye laser radiation.

Apart from the tuning range, other facts of importance is the intensity of the generated radiation that is necessary

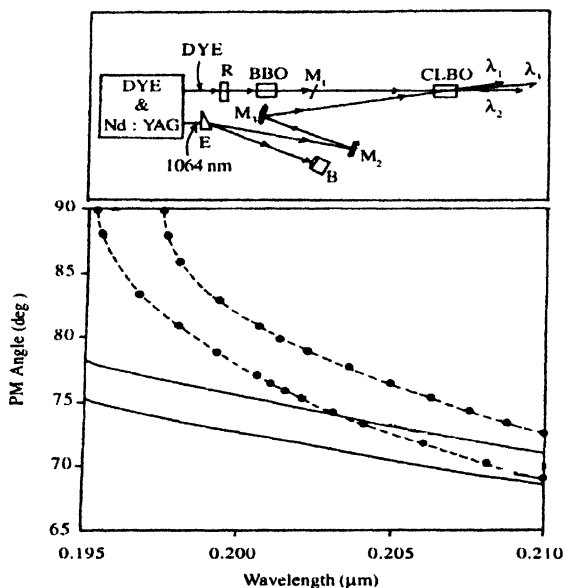


Figure 3. Phase-matching characteristics of CLBO crystal for generation of 195.3–210 nm by noncollinear SFM of Nd : YAG (1064 nm) and dye SHG (239.2–262 nm) radiations for internal noncollinear angle $\alpha = 0.8^\circ$ (lower dashed curve) and 8° (upper dashed curve). Lower and upper solid curves are theoretically predicted ones for $\alpha = 0.8^\circ$ and 8° respectively. The inset shows the schematic of the experimental setup. M_1 is a dichoric mirror transmitting only the generated SHG (239.2–262 nm). M_2 and M_3 are also dichoric mirrors for 1064 nm. B and E are respectively beam dumper and extra dense flint glass prism. R is a 90° polarization rotator for dye laser radiation. The BBO crystal is rotated in a vertical plane while the CLBO in horizontal plane to satisfy proper phase-matching conditions.

to deactivate the microorganism. This is tied up with the frequency conversion efficiency in the nonlinear process. While the tuning wavelength is dictated by the crystal phase-matching properties, for the efficiency of conversion, we have to look for ingenious techniques other than crystal properties. Two such techniques for increasing efficiency have been demonstrated in the laboratory with success :

- (i) Walk-off compensation (WOC) configuration,
- (ii) Multipass configuration (MPC).

Birefringently phase-matched frequency conversion process occurs in presence of both ordinary and extraordinary polarized radiations. The later while propagating through the crystal, suffers birefringent walk-off whereas the former is not. As a result of separation of the beams, the effective crystal interaction length is limited which reduces conversion efficiency. To overcome this limitation, it is suggested that the walk-off produced in one crystal is to be compensated by using another crystal with its optic

axis in other direction as shown in Figure 4. Such a crystal pair of BBO is employed for THG of dye laser radiation 200–230 nm [6]. We demonstrate 36% conversion efficiency for UV at 210 nm. In LB4 crystal pair, we have generated 288 nm UV radiation at power level of 21 MW by the same technique.

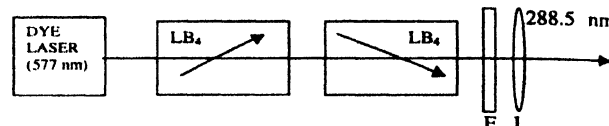


Figure 4. The experimental setup for WOC SHG scheme in LB4 crystals. L is CaF_2 lens. F is dichoric mirror transmitting $>70\%$ at 288.5 nm only.

Still more advantageous technique is multipass configuration illustrated in Figure 5 where twin crystal of WOC set up is ingeniously used for more than one pass through the pair and hence the nonlinear conversion is further improved [7]. With this it has been possible to realize 21% conversion efficiency in LB4 crystal in SHG of Nd : YAG laser radiation, even though the nonlinearity of LB4 is 1/6-th that of KDP.

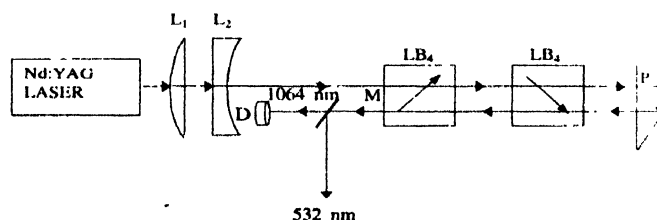


Figure 5. Generation of 532 nm by SHG of Nd : YAG laser radiation using multipass configuration in LB4 crystals. L_1 and L_2 are lenses. P is prism and D is a beam dumper for Nd : YAG laser radiation. Dichoric mirror M reflects $>98\%$ at 532 nm and transmits about 100% of 1064 nm radiation.

4. Conclusion

In conclusion, we have demonstrated high intensity, widely tunable UV source capable of deactivation/removal of even highly resistant protozoan cysts using commercially available Nd : YAG and Dye laser and common nonlinear crystals.

Acknowledgments

The authors acknowledge the partial financial support from the DRDO, Ministry of Defence, Government of India. They are also grateful to the authorities of VECC, Kolkata, Government of India for input laser power supply control and stability.

References

- [1] M J Parolta and F Beckdash *J. Am. Water Work Assn.* **90** 71 (1998)
- [2] R L Wolfe *Enviorn. Sci. Technol.* **24** 768 (1990)
- [3] G C Bhar, U Chatterjee, A M Rudra, P Kumbhakar, R K Route and R S Feigelson *Opt. Lett.* **22** 1606 (1997)
- [4] H Kouta and Y Kuwano *Opt. Lett.* **24** 1230 (1999)
- [5] G C Bhar, P Kumbhakar, U Chatterjee, A K Chaudhary and A Kokh *Pramana* **59** 69 (2002)
- [6] U Chatterjee, S Gangopadhyay, C Ghosh, P Chattopadhyay and G C Bhar *Jpn. J. Appl. Phys.* **43** 4190 (2004)
- [7] U Chatterjee, S Gangopadhyay, C Ghosh and G C Bhar *Appl. Opt.* **44** 817 (2005)